

PCT/CA 03/00593

JUNE 2003 09 • 06 • 03

PA 1016184

RECD 19 JUN 2003

WIPO PCT

# THE UNITED STATES OF AMERICA

TO ALL TO WHOM THESE PRESENTS SHALL COME:

UNITED STATES DEPARTMENT OF COMMERCE

United States Patent and Trademark Office

May 29, 2003

THIS IS TO CERTIFY THAT ANNEXED HERETO IS A TRUE COPY FROM  
THE RECORDS OF THE UNITED STATES PATENT AND TRADEMARK  
OFFICE OF THOSE PAPERS OF THE BELOW IDENTIFIED PATENT  
APPLICATION THAT MET THE REQUIREMENTS TO BE GRANTED A  
FILING DATE UNDER 35 USC 111.

APPLICATION NUMBER: 60/374,512  
FILING DATE: April 23, 2002

BEST AVAILABLE COPY

**PRIORITY  
DOCUMENT**

SUBMITTED OR TRANSMITTED IN  
COMPLIANCE WITH RULE 17.1(a) OR (b)

By Authority of the  
COMMISSIONER OF PATENTS AND TRADEMARKS



  
N. WILLIAMS  
Certifying Officer

**PROVISIONAL APPLICATION FOR PATENT COVER SHEET**

This is a request for filing a PROVISIONAL APPLICATION FOR PATENT under 37 CFR 1.53(c).

Express Mail Label No. \_\_\_\_\_

**INVENTOR(S)**

Given Name (first and middle [if any])	Family Name or Surname	Residence (City and either State or Foreign Country)
LORRIE	HUNT	Delta, Canada
JIM	McKINLEY	North Vancouver, Canada
ROD	McELROY	

 Additional inventors are being named on the \_\_\_\_\_ separately numbered sheets attached hereto**TITLE OF THE INVENTION (500 characters max)****REMEDIATION OF SOILS CONTAMINATED WITH POLYCHLORINATED BIPHENYLS (PCBs) AND OTHER ORGANOHALOGENS BY SIMULTANEOUS PCB EXTRACTION AND DESTRUCTION**

Direct all correspondence to:		CORRESPONDENCE ADDRESS			
<input type="checkbox"/>	Customer Number	<input type="text"/>			
OR		<input type="text"/> Place Customer Number Bar Code Label here			
<input checked="" type="checkbox"/>	Firm or Individual Name	Vermette & Co.			
Address	Box 40, Granville Square				
Address	Suite 230, 200 Granville Street				
City	Vancouver	State	British Columbia	ZIP	V6C 1S4
Country	Canada	Telephone	(604) 331-0381	Fax	(604) 331-0382

**ENCLOSED APPLICATION PARTS (check all that apply)**

<input checked="" type="checkbox"/> Specification	Number of Pages	<input type="text"/> 0	<input type="checkbox"/> CD(s), Number	<input type="text"/>
<input type="checkbox"/> Drawing(s)	Number of Sheets	<input type="text"/>	<input type="checkbox"/> Other (specify)	<input type="text"/>
<input type="checkbox"/> Application Data Sheet. See 37 CFR 1.76				

**METHOD OF PAYMENT OF FILING FEES FOR THIS PROVISIONAL APPLICATION FOR PATENT**

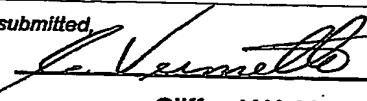
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27.	FILING FEE AMOUNT (\$)
<input type="checkbox"/> A check or money order is enclosed to cover the filing fees	
<input type="checkbox"/> The Commissioner is hereby authorized to charge filing fees or credit any overpayment to Deposit Account Number: <input type="text"/>	
<input checked="" type="checkbox"/> Payment by credit card. Form PTO-2038 is attached.	

\$160.00

The invention was made by an agency of the United States Government or under a contract with an agency of the United States Government.

 No. Yes, the name of the U.S. Government agency and the Government contract number are: \_\_\_\_\_

Respectfully submitted,

SIGNATURE Date 

TYPED or PRINTED NAME Clifford W. Vermette

REGISTRATION NO.  
(if appropriate)  
Docket Number:

TELEPHONE (604) 331-0381

30,018

1635-255

**USE ONLY FOR FILING A PROVISIONAL APPLICATION FOR PATENT**

This collection of information is required by 37 CFR 1.51. The information is used by the public to file (and by the PTO to process) a provisional application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 8 hours to complete, including gathering, preparing, and submitting the complete provisional application to the PTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, Washington, D.C. 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Box Provisional Application, Assistant Commissioner for Patents, Washington, D.C. 20231.

Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

# FEE TRANSMITTAL

## for FY 2002

Patent fees are subject to annual revision.

TOTAL AMOUNT OF PAYMENT (\$ 160.00)

**Complete if Known**

Application Number	
Filing Date	
First Named Inventor	Lorrie Hunt
Examiner Name	
Group Art Unit	
Attorney Docket No.	2313-100

**METHOD OF PAYMENT**

1.  The Commissioner is hereby authorized to charge indicated fees and credit any overpayments to:
- Deposit Account Number
- Deposit Account Name
- Charge Any Additional Fee Required Under 37 CFR 1.16 and 1.17
- Applicant claims small entity status. See 37 CFR 1.27

2.  Payment Enclosed:
- Check  Credit card  Money Order  Other

**FEE CALCULATION****1. BASIC FILING FEE**

Large Entity Small Entity

Fee Code	Fee (\$)	Fee Code	Fee (\$)	Fee Description
101	740	201	370	Utility filing fee
106	330	206	165	Design filing fee
107	510	207	255	Plant filing fee
108	740	208	370	Reissue filing fee
114	160	214	80	Provisional filing fee

Fee Paid  
  
  
  
  
  
160.00

SUBTOTAL (1) (\$ 160.00)

**2. EXTRA CLAIM FEES**

Total Claims	3	Extra Claims	Fee from below	Fee Paid
Independent Claims		-20** = <input type="text"/>	X <input type="text"/>	= <input type="text"/>
Multiple Dependent		- 3** = <input type="text"/>	X <input type="text"/>	= <input type="text"/>

## Large Entity Small Entity

Fee Code	Fee (\$)	Fee Code	Fee (\$)	Fee Description
103	18	203	9	Claims in excess of 20
102	84	202	42	Independent claims in excess of 3
104	280	204	140	Multiple dependent claim, if not paid
109	84	209	42	** Reissue independent claims over original patent
110	18	210	9	** Reissue claims in excess of 20 and over original patent

SUBTOTAL (2) (\$ )

\*\*or number previously paid, if greater; For Reissues, see above

**FEE CALCULATION (continued)****3. ADDITIONAL FEES**

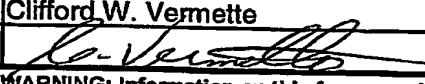
Fee Code	Large Entity Fee (\$)	Small Entity Fee Code (\$)	Fee Description	Fee Paid
105	130	205	65 Surcharge - late filing fee or oath	<input type="text"/>
127	50	227	25 Surcharge - late provisional filing fee or cover sheet	<input type="text"/>
139	130	139	130 Non-English specification	<input type="text"/>
147	2,520	147	2,520 For filing a request for ex parte reexamination	<input type="text"/>
112	920*	112	920* Requesting publication of SIR prior to Examiner action	<input type="text"/>
113	1,840*	113	1,840* Requesting publication of SIR after Examiner action	<input type="text"/>
115	110	215	55 Extension for reply within first month	<input type="text"/>
116	400	216	200 Extension for reply within second month	<input type="text"/>
117	920	217	460 Extension for reply within third month	<input type="text"/>
118	1,440	218	720 Extension for reply within fourth month	<input type="text"/>
128	1,960	228	980 Extension for reply within fifth month	<input type="text"/>
119	320	219	160 Notice of Appeal	<input type="text"/>
120	320	220	160 Filing a brief in support of an appeal	<input type="text"/>
121	280	221	140 Request for oral hearing	<input type="text"/>
138	1,510	138	1,510 Petition to institute a public use proceeding	<input type="text"/>
140	110	240	55 Petition to revive - unavoidable	<input type="text"/>
141	1,280	241	640 Petition to revive - unintentional	<input type="text"/>
142	1,280	242	640 Utility issue fee (or reissue)	<input type="text"/>
143	480	243	230 Design issue fee	<input type="text"/>
144	620	244	310 Plant issue fee	<input type="text"/>
122	130	122	130 Petitions to the Commissioner	<input type="text"/>
123	50	123	50 Processing fee under 37 CFR 1.17(q)	<input type="text"/>
126	180	126	180 Submission of Information Disclosure Stmt	<input type="text"/>
581	40	581	40 Recording each patent assignment per property (times number of properties)	<input type="text"/>
146	740	246	370 Filing a submission after final rejection (37 CFR § 1.129(a))	<input type="text"/>
149	740	249	370 For each additional invention to be examined (37 CFR § 1.129(b))	<input type="text"/>
179	740	279	370 Request for Continued Examination (RCE)	<input type="text"/>
169	800	169	800 Request for expedited examination of a design application	<input type="text"/>

Other fee (specify) \_\_\_\_\_

\*Reduced by Basic Filing Fee Paid

SUBTOTAL (3) (\$ )

**SUBMITTED BY**

Name (Print/Type)	Clifford W. Vermette	Registration No. (Attorney/Agent)	30,018	Telephone	(604) 331-0381
Signature					
Date	03/14/2002				

**WARNING:** Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.

Burden Hour Statement: This form is estimated to take 0.2 hours to complete. Time will vary depending upon the needs of the individual case. Any comments on 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Assistant Commissioner for Patents, Washington, DC 20231.

**REMEDIATION OF SOILS CONTAMINATED WITH POLYCHLORINATED BIPHENYLS  
(PCBs) AND OTHER ORGANOHALOGENS BY SIMULTANEOUS PCB EXTRACTION  
AND DESTRUCTION**

**5 FIELD**

The present invention relates to a method of simultaneously extracting and destroying polychlorinated biphenyls (PCBs) in contaminated solids, such as soil.

**10 BACKGROUND**

The existence of large numbers of sites with soil extensively contaminated by organohalogens, especially PCBs, requires economical cost effective treatment methods. Although incineration has been shown capable of destroying PCBs this form of PCB treatment has been implicated in dioxin and chlorinated dibenzofuran emissions and has been banned for use in certain countries such as Australia and Japan (References 1,2). Both furan and dioxins are considered to be highly toxic (carcinogenic). As such, there is a need for cost effective PCB decontamination methods, including soil decontamination, which do not involve PCB incineration.

Getman et al in U.S. patent 6,049,021 describe remediation of soil contaminated with PCBs. This patent describes the destruction of PCBs in soil using a variety of methods combining the following basic elements:

1. PCB extraction of soil by liquid ammonia.

2. Dissolution of sodium metal into PCB-contaminated liquid ammonia.

5           3. Destruction of PCB in liquid ammonia by dissolved sodium metal.

Although this technique clearly shows high destruction of PCB in soil it suffers from the following problems:

- 10         • Very long reaction times e.g. >30 minutes (see Example 2).
- 15         • Need to refrigerate ammonia with soil with stirring before addition of sodium metal (see Example 4).
- 20         • Need to operate with hazardous pressurized anhydrous ammonia gas in a stirred vessel (see Example 2).
- 25         • Extremely high ammonia dose on soil e.g. 9 litres ammonia per kilogram of soil (see Example 2).
- 30         • Generation of ammonia containing residual wastes "filtrates" (see Example 2).
- 35         • Awkward temperature cycling between 0 °C and 20 to 40 °C (see Example 4) or -78°C (see Example 3).
- 40         • Awkward, time consuming, multiple soil extractions with ammonia before addition of sodium metal (see Example 4).

United States patent 5,228,921 to Peterson describes a method for extracting organohalogens from organohalogen contaminated solids such as PCBs from PCB contaminated soils.

United States patent 5,376,182 to Everett et al describe PCB extraction from PCB contaminated soil with ultrasound at 10 to 5 60 kilohertz frequency. Although these extraction methods successfully remove PCBs from soil, they do not destroy the PCBs.

PCT application WO 02/22252 A1 to Collings describes ultrasonic destruction of PCBs in a one-step process. However processing times are excessive (e.g. 60 minutes) and PCB destruction efficiency is low (e.g. 75%) (page 10, lines 20-25).

Eco Logic at web page [www.eco-logic-intl.com](http://www.eco-logic-intl.com) in a brochure dated April 2001 and entitled "The TORBED/GPCR combination for Soil, Sediment and Sludge Treatment" describe a multi-step process for removal and destruction of PCBs in solids such as soils as follows:

1. High temperature (e.g. 600°C) thermal desorption of PCBs from soils by volatilization.
2. High temperature (e.g. 875°C) gas phase reduction of volatilized PCB exhaust gas from step 1 with a reducing gas such as hydrogen.
3. Scrubbing of exhaust gas from step 2 to recover toxic and/or corrosive gases such as hydrogen chloride produced from reduction of PCBs.

4. Compression and/or storage of scrubbed exhaust gas from step 3.
5. Incineration and/or recycling of scrubbed exhaust gas from step 4 to steps 1 and/or 2 respectively.

Although the Eco Logic method clearly destroys PCBs in soils it suffers from the following problems:

SEARCHED  
SERIALIZED  
INDEXED  
FILED

10

- Generation of toxic and/or corrosive exhaust gas (e.g. hydrogen chloride) and spent scrubber solutions.
- Use of potentially explosive hydrogen gas at high temperature.
- Five or more processing steps.
- Two energy intensive, high temperature processing steps.

20

There is a need for a low temperature process which can quickly extract and destroy PCBs in one step thereby reducing the number of processing steps, the size of equipment and their cost.

25 **SUMMARY OF THE INVENTION**

The current invention relates to the simultaneous extraction and destruction of PCBs in contaminated solids, such as soil. The current invention quickly extracts and destroys PCBs by combining the contaminated soil with an alkali metal,

such as sodium, as well as an inert alkali-metal liquid carrier, such as kerosene. The combination is then placed in a sealed sonication chamber to effect melting of the alkali metal, which causes simultaneous extraction of PCB from the contaminated soil  
5 into the liquid carrier and destruction of PCBs by the molten alkali metal. A method of stripping the inert alkali-metal liquid carrier is then employed, resulting in remediation of the contaminated soil.

10 In contrast to the prior art, the use of this method allows remediation of solids, such as soil, in one reaction chamber using sonication, and at relatively low temperatures, which avoids the highly toxic emissions caused by high temperature incineration.

15 One advantage of the current invention is the use of kerosene as an extractant resulting in a significant reduction in the amount of extractant required.

20 **DETAILED DESCRIPTION**

The following remediation method can be applied to solids, such as soil, contaminated by organohalogens, especially PCBs:

25 1. Air-dry and sieve PCB contaminated soil.

2. Combine sieved, air-dried PCB contaminated soil from step 1 with an alkali metal (e.g. sodium) in an essentially inert alkali-metal liquid carrier (e.g. kerosene or other hydrocarbon) and place inside a sealed sonication chamber.

30

3. Heat the sonication chamber from step 2 to effect melting  
of the alkali-metal (e.g. sodium) and apply sonication to  
effect simultaneous extraction of PCB from the contaminated  
solids into the liquid carrier and destruction of extracted  
PCBs by molten alkali metal (e.g. sodium).

5

4. Discharge the sonication chamber reaction product from step  
3 and use any one of the non-limiting soil/liquid carrier  
separation, alkali hydroxide (e.g. sodium hydroxide) by-  
product neutralization methods described below.

10

5. Recycle the treated soil.

Referring to step 4, a variety of non limiting-techniques  
15 for stripping the inert alkali-metal carrier (e.g. hydrocarbon  
such as kerosene) from the soil together with soil recycling  
include the following:

20

25

- For coarse-grained soils, addition of water (the same or higher volume than the hydrocarbon oil) followed by vigorous mixing in the sonic generator chamber (or in a separate vessel) results in water wetting of the solid phase. When the agitation is stopped, the oil floats on the water phase and can be recovered by decantation. Water addition/mixing followed by settling and oil decantation will be the primary stage of oil stripping in most cases.
  
- More commonly the soil in the water phase after primary oil decantation will retain a significant amount of oil and

require treatment by froth flotation to achieve the required oil content for recycling.

- When stringent criteria (<20 ppm) apply to the oil content of recycle soil, oil sorption by coarse granular activated carbon can be used to achieve very low residual oil levels.

- 5
- The oil recovered by decantation will normally contain a portion of the fine-grained solids from the treated soil.

10 Depending on the nature of the soil being treated, methods to recover solids from the recycle oil phase include secondary water scrubbing with a surfactant, centrifugation or filtration.

- 15
- Removal of water from treated soil can be achieved by simple drainage for coarse-grained soil. For finer material vacuum or pressure filtration will provide a handleable material for recycle.

20 The methods described above are either standard practice or slight variants of standard mineral processing operations (references 3,4).

25 Modern practice in solvent extraction also provides examples of technology for removal of fine solids ("crud" in industry jargon) from the organic phase which typically contains >50% hydrocarbon oil as diluent as well as the oil soluble extraction reagent.

During the water contact /oil separation operations, sodium hydroxide (NaOH) is formed from any residual sodium metal and by hydrolysis of the sodium-aromatic hydrocarbon bond formed during the dechlorination reaction. Sodium hydroxide is known in the

5 mineral processing industry as a (water) wetting agent for many minerals, and may assist in the oil removal operation. However, neutralization of sodium hydroxide will be required prior to discharge of process water. This can be achieved by a variety of non-limiting methods including:

- 10 • Injection of carbon dioxide gas into the aqueous phase to convert sodium hydroxide into sodium carbonate or bicarbonate  
• Addition, under pH control, of sulfuric acid to form neutral sodium sulphate

15 The following example illustrates the effectiveness of the invention:

**Example 1**

1. PCB-contaminated soil was air dried and sieved to -6 mesh.

20 2. Two kilograms of soil from step 1 was combined with 0.6 litres of commercially available kerosene (i.e. a 0.3 litre kerosene to 1 kg soil weight ratio) and 45 grams of solid sodium metal in a 3.2 litre grinding media-free sonication chamber attached to a 20 kilowatt sonic driver.

25 3. The sealed sonication chamber was mounted on the 20 kilowatt sonic driver and was heated to 115°C using heat from a propane torch. The sonic chamber heating jacket was filled half-

way with ethylene glycol antifreeze to aid in heat transfer to the sonic chamber ingredients. Once the desired temperature was reached (e.g. alkali metal melting point or higher), sonication was commenced.

5

As an example, the sonication temperature was 114°C at the end of a 6 minute run. The following table illustrates PCB destruction as a function of time using the above approach on a soil with an initial PCB content of 424 ppm (micrograms/gram):

10

Sonication Time in minutes	PCB ppm in soil	% PCB destruction
0	424	0
1	12.7	97.0
2	8.4	98.0
5	2.2	99.5
6	2.5	99.4

A 99.5% destruction of 424 ppm PCB contaminated solid with a 45 g. sodium to 2000 g soil dose ratio was achieved in 5 minutes.

15

Using kerosene as an extractant required approximately 0.3 liters of kerosene to one kilogram of soil. This ratio represents a 30 fold reduction in the volume of extractant required as compared to the ammonia extractant of the Getman patent.

20

**References**

1. Costner, Pat et al. 1998. "Technical Criteria for the  
Destruction of Stockpiled Persistent Organic Pollutants".  
5 Third Meeting of the Intersessional Group Intergovernmental  
Forum on Chemical Safety. Yokohama, Japan. December 1-4,  
1998. Available at [www.eco-logic-intl.com](http://www.eco-logic-intl.com)
- 10 2. "Survey of Currently Available Non-Incineration PCB  
Destruction Technologies". 2000. United Nations Environment  
Programme. August. Available at [www.eco-logic-intl.com](http://www.eco-logic-intl.com)
- 15 3. Taggart, Arthur F. 1945. Handbook of Mineral Dressing. John  
Wiley and Sons Inc. (New York)
4. Gaudin, A.M. 1957. Flotation. McGraw-Hill Book Co. Inc.  
(New York)